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(54) Alkyl ether mono amines.

(57) Distillate fuel compositions containing an alkyl ether mono amine preferably derived from a Guerbet alcohol are effective in reducing the formation of intake valve deposits in internal combustion engines. A preferred alkyl ether mono amine is derived from a highly branched butoxylated Guerbet alcohol containing 20 carbon atoms and having the formula



where R is a highly branched alkyl group having 20 carbon atoms that is derived from a Guerbet condensation reaction.

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ALKYL ETHER MONO AMINES

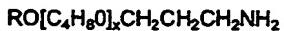
This invention concerns alkyl ether mono amines and their use in distillate fuels to reduce the formation of intake valve deposits in an internal combustion engine.

The use of alkyl ether mono and polyamines is known. For example, U.S. Patent 3,440,029 discloses a broad class of alkyl ether mono amines and their use as gasoline deicing additives in carbureted vehicles. However, although these alkyl ether mono amines may be effective deicers, many (if not most) of them are ineffective gasoline intake system detergents. Polyamines are also disclosed in U.S. Patents 4,247,301; 4,332,595; and 4,604,103.

In addition, European Patent Application 310,875 discloses the use of certain polyether mono amines prepared by the reductive amination of certain alcohols with ammonia or primary aliphatic amines. Similarly, European Patent Applications 181,140 and 180,455 disclose the use of certain polyether tertiary amines.

However, none of these publications concern alkyl ether mono amines derived from Guerbet alcohols nor the use of these amines in the fuel of an internal combustion engine.

This invention concerns a particular class of alkyl ether mono amines and their use in a distillate fuel. More specifically, the invention provides a distillate fuel composition comprising (a) a gasoline, and (b) an alkyl ether mono amine having the formula:



where

R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and

x is from 0 to 30.

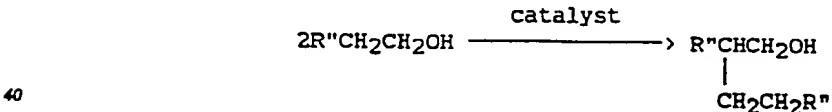
We have discovered that a fuel containing a major amount of gasoline and a minor amount of an oil soluble alkyl ether mono amine as defined above can reduce the formation of intake valve deposits in an internal combustion engine. A fuel containing these amines can also reduce fuel injector deposits in a fuel injected internal combustion engine. The alkyl ether mono amine is preferably derived from a Guerbet alcohol, especially a highly branched butoxylated Guerbet alcohol. In a particularly preferred embodiment, the alkyl ether mono amine is derived from a highly branched butoxylated Guerbet alcohol containing 20 carbon atoms and having the following formula:



where R' is a highly branched alkyl group having 20 carbon atoms that is derived from a Guerbet condensation reaction.

The Guerbet reaction was first described in 1899 by M. Guerbet as a method for condensing two small alcohols into a larger branched alcohol, wherein the branch point occurs at the "beta" carbon (see C.R. Acad. Sci. Paris, 128,511;1002). However, many refinements to the original method of preparation have occurred [e.g. Tetrahedron, vol. 23, page 1723, (1967)]. The overall Guerbet reaction can be represented as follows:

35



40 where R'' is a hydrocarbyl chain. The product of this reaction is an alcohol with twice the molecular weight of the reactant alcohol minus a mole of water. Much is known about the complex sequence of reactions which comprises the overall reaction shown above. The mechanism is extensively discussed in Tetrahedron, supra. Many catalysts have been described in the literature as being effective for preparing Guerbet alcohols. These catalysts include nickel; lead salts (U.S. patent 3,119,880); oxides of copper, lead, zinc, chromium, molybdenum, tungsten, and manganese (U.S. Patent 3,558,716); palladium compounds and silver compounds (U.S. Patent 3,864,407).

45 Guerbet alcohols have unusual properties. These unique properties are partly attributed to their high molecular weight and high level of saturation (A. J. O'lenick Jr. and R. E. Bilbo, Soap/Cosmetics/ Chemical Specialties, April 1987, page 52). Unusual properties are also attributed to the so called "beta branch point" (presentation by R. Varadaraj et al., at the American Oil Chemists Society Meeting in Cincinnati, Ohio during May 3-6, 1989). Some of the properties attributed to Guerbet alcohols are low irritation, liquidity to extremely 50 low temperatures, low volatility, relatively reactive and easy to derivatize, useful superfatting agents to re-oil the skin and hair, highly lipophilic, good oxidation stability, and excellent color stability. However, most laborat-

ory studies and commercial applications of Guerbet alcohols and their derivatives have utilized water based systems. Very little, if any, published information is available on their use in hydrocarbon based applications such as gasoline or distillate fuels.

The preferred alkyl ether mono amines for use in the distillate fuel composition of this invention are Guerbet alkyl ether mono amines which are oil soluble and have the general formula:



wherein:

R is a highly branched alkyl group derived from a Guerbet alcohol containing between 12 and 40 carbon atoms. By "highly branched" is generally meant that R contains at least four substituent methyl groups; and

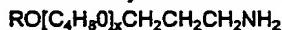
x is the number of moles of butylene oxide, which may range from 0 to 30, preferably from 1 to 30, more preferably from 1 to 20, even more preferably from 1 to 10, and especially from 4 to 8.

More specifically, R preferably has the general formula $R_1R_2CHCH_2$ wherein R_1 contains from 6 to 20 (preferably from 8 to 15) carbon atoms and R_2 contains from 4 to 18 (preferably from 6 to 13) carbon atoms. In the most preferred embodiment, R is a Guerbet derived, highly branched alkyl group containing about 20 carbon atoms and x is about 8.

In general, the molecular weight of the Guerbet ether amines will not be monodisperse. The process used for adding the butylene oxide will generally result in a distribution of moles of butylene oxide centered around the average x, which may or may not be an integer.

The distillate fuel composition of this invention will, in general, comprise a major amount of gasoline and a minor amount of the highly branched Guerbet ether mono amines described above. However, the precise amount of Guerbet ether amine can vary broadly. As such, only an amount effective or sufficient to reduce the formation of intake valve deposits or fuel injector deposits need be used. Typically, however, the amount of Guerbet ether amine used will range from about 50 to about 2000 ppm, although greater amounts could be used. Preferably, from about 50 to 1000, more preferably from about 100 to about 500, ppm of Guerbet ether amine will be present in the fuel.

Certain of the Guerbet alkyl ether mono amines disclosed herein are novel compounds per se. Thus in another aspect the present invention provides an alkyl ether mono amine having the formula:



where

R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and
x is from 1 to 30.

The preferred definitions given above for R and x in the alkyl mono ether amine incorporated into the distillate fuel composition also apply to R and x of the above formula.

The Guerbet alkyl ether mono amines of this invention may be readily prepared by methods known in the art. They may most conveniently be prepared by reaction of the Guerbet alcohol with butylene oxide using a base catalyst at a temperature of 250° to 375°F. The relative concentration of Guerbet alcohol to butylene oxide will depend on the final product desired. The resulting Guerbet ether alcohol can then be reacted with acrylonitrile, in the presence of alkali at 40° to 135°F, to produce the Guerbet ether nitrile. This reaction is usually carried out with equal moles of the Guerbet ether alcohol and acrylonitrile, although a small excess of acrylonitrile can be used to increase the degree of reaction. The reaction product is then filtered to remove excess polyacrylonitrile. The Guerbet ether nitrile is then reduced in the presence of hydrogen, ammonia, and catalyst, at a temperature of 200° to 350°F, to produce the Guerbet ether mono amine.

Although the Guerbet alkyl ether mono amines used herein will generally be added to distillate fuel, they may be formulated as a concentrate using a hydrocarbon solvent, an alcohol solvent, or mixtures thereof boiling in the range of about 150 to about 400°F. Preferably, an aromatic hydrocarbon solvent (such as benzene, toluene, xylene, or higher boiling aromatics or aromatic thinners, and the like) is used. Aliphatic alcohols of about 3 to 8 carbon atoms (such as isopropanol, isobutylcarbinol, n-butanol, and the like), alone or in combination with hydrocarbon solvents, can also be used with the Guerbet alkyl ether mono amines. The amount of Guerbet alkyl ether mono amine in the concentrate will ordinarily be at least about 10 wt. % and, generally, will not exceed about 70 wt. %. Similarly, the amount of hydrocarbon solvent will typically range from about 30 to about 90 wt. % of the concentrate.

The distillate fuel composition of this invention (including the fuel concentrate) may also contain a small amount (typically from about 0.02 to about 0.5 wt. % and preferably from about 0.02 to about 0.15 wt. %) of a carrier fluid of low volatility. As used herein, the term "carrier fluid" is meant to include hydrocarbon and oxygenated species. Typically, the carrier fluid will have a kinematic viscosity of between 4 and 75 cSt at 100°C. Examples of such carrier fluids include lubricating oil base stocks, alcohols, polyols, polyesters, polyalkylene oxides (e.g. Ucon Fluids available from Union Carbide), their mixtures and the like. Sometimes these carrier fluids demonstrate synergistic intake system detergency when used in combination with the Guerbet eth

mono amines of this invention.

Other additives may be included in the fuel. Examples of such additives include antiknock agents (e.g. tetraethyl lead), other detergents or dispersants, demulsifiers, antioxidants, anticorrosives, and the like.

Although the Guerbet alkyl ether mono amines of this invention are particularly effective intake valve detergents, these amines are also effective in reducing fuel injector deposits in fuel injected internal combustion engines, especially multipoint electronically controlled fuel injected engines.

This invention will be further understood by reference to the following Examples which are not intended to limit the scope of the claims appended hereto.

10 Example 1 - Preparation of C₂₆ Guerbet Ether Mono Amines

A. Preparation of Poly(oxybutylene)monool of a C₂₆ Guerbet alcohol

Exxal 26 (2430 g, 5.63 mols), a C₂₆ Guerbet alcohol available from Exxon Chemical Company, and a solution of potassium hydroxide (KOH) in water (45% by weight; 23.64 g, 10.64 g active KOH or 0.20 mol; 0.25 weight %, based on total reactor charge) were introduced into a standard bottom discharge 2 gallon T316 stainless steel Autoclave Engineers' high pressure reactor, equipped with a 600 psi pressure gauge, cooling coils, 1000 psi rupture disc, and a vacuum distillation take-off adapter. While applying a vacuum of 25 to 26 in. Hg to the system, the contents of the reactor were stirred and heated to 112°C for 2 hours until no residual water was seen to condense on the condensor. The contents were then cooled to ambient temperature (25°C) by cooling the reactor with cold water several minutes via the cooling coil. 1,2-epoxybutane (1828 g, 25.38 mols or 4.5 mols of alkylene oxide/mol Exxal 26) was then charged to the reactor through a port opened on the top of the reactor. The port was sealed and the valve leading to the distillation take-off was closed. The mixture was heated to 170° to 175°C over two hours during which time the pressure rose to 82 psi. The pressure then dropped steadily to 0 psi over 45 minutes as the reaction proceeded. When the pressure reached 0 psi, heating was continued at the same temperature for an additional hour to ensure completion. The contents were then cooled to 120°C, the valve to the distillation take-off opened, and vacuum of 25 to 26 in. Hg applied over a half hour to remove any unreacted butylene oxide. The reaction mixture was then cooled to 90°C and vacuum filtered hot through a bed of filter aid. 3763.1 g of an amber colored liquid were collected (88.5% of theoretical).

30 B. Preparation of C₂₆ Guerbet poly(oxybutyl)-oxypropanonitrile

The butoxylated alcohol prepared above (4.98 mol) was introduced to a 5 liter 4-neck round bottom flask equipped with a thermometer, overhead stirrer, condensor, and a dropping funnel. A few drops of 45 wt% KOH in water were added to catalyze the reaction. The contents of the flask were heated to 30°C using a heating mantle with stirring. Acrylonitrile (383 g, 7.21 mol) was charged to the dropping funnel and approximately 50 ml aliquots were added in a fast stream over a couple of minutes at about 15 to 20 minute intervals initially over 2.5 hours in such a manner that maintained the temperature at less than 40°C. After adding 275 g of the acrylonitrile, an additional 3 g of 45 wt% KOH in water was added. The last 108 g of acrylonitrile was added over the next 6 hours, in 20 to 30 g aliquots while monitoring the nitrile and hydroxyl functionality by infrared spectroscopy. The mixture was stirred an additional 2 hours until the infrared spectrum showed no further conversion of hydroxyl functionality. Approximately 70% of the hydroxyl groups had been reacted according to the infrared analysis. 25 ml of water was then added. The mixture was allowed to sit an hour at 40°C and the water settled. 0.5 N hydrochloric acid was then added dropwise with stirring until the pH of the reaction mixture was neutral (according to pH paper). The neutralized solution was poured through a large filter funnel through #1 Whatman filter paper to remove any acrylonitrile polymer and inorganic salts. 3653 g of the amber colored filtrate (90.7% of theoretical) of the ether nitrile was isolated.

C. Preparation of C₂₆ Guerbet poly(oxybutyl)-oxypropylamine

50 Raney Nickel catalyst (200.9 g, 5.5 wt%, based on ether nitrile) was washed 3 times with 500 ml aliquots of isopropanol. In the first two cases, the solvent was decanted off and fresh solvent added. After suspending the catalyst in the third aliquot of isopropanol, the mixture was added to the 2 gallon Autoclave Engineers' reactor described above. The ether nitrile prepared in B above (3653 g, 4.52 mol) was then added to the reactor and stirring begun. A vacuum of 25 in. Hg was applied to the system by opening the valve of the distillation take-off and the contents of the reactor heated to 120°C. The isopropanol and any residual water was removed by distillation over 2 hours until no condensate was seen forming on the condensor. The distillation valve was closed and the reactor sealed. Hydrogen was then added to a pressure of 10 psi and the reactor vented. The

hydrogen purge and venting were repeated. Hydrogen was again added to a pressure of 10 psi and the contents of the reactor cooled over a few minutes to 70°C by admitting cold water through the cooling coils. Ammonia was added to raise the pressure to 100 psi. Heating was continued and the temperature of the contents increased to 135°C (pressure had increased to 160 psi) over approximately 30 minutes. Hydrogen was added 5 to maintain the total pressure at 320 psi and the temperature was maintained at 135° to 140°C for 32 hours. A small sample was taken from a sample port and analyzed for completion of reaction by titrimetric methods and by Infrared Spectrophotometry. The contents of the reactor were cooled to 120°C, vented, and a vacuum of 10 25 in. Hg applied by opening the distillation valve. The contents were then vacuum distilled for 2 hours to remove residual ammonia. The contents were further cooled to 50°C, then drained from the bottom discharge valve of the reactor, and vacuum filtered warm through a bed of filter aid. In this manner 3240.6 (88.3% of theoretical) 10 were isolated. An additional 200 to 400 g could be isolated by extraction of the filter aid and catalyst by slurring in a liter of hexane, followed by refiltration and concentration of product by distillation of the solvent. The yields thus approached 93-99% of theoretical.

15 Example 2 - Performance of Highly Branched Guerbet Ether Mono Amines Derived from Butylene Oxide

Highly branched C₂₀ and C₂₆ Guerbet ether mono amines were synthesized as described in Example 1 from butylene oxide and then blended (at various concentrations) into two different commercial unleaded 93 octane base gasolines. Both gasolines contained small amounts of antirust and antioxidant stabilizers. (Unless 20 otherwise stated, the 93 octane gasolines used in the other examples also contained these stabilizers.) The fuels were then tested in a BMW 325 for 100 hours on a standard mileage accumulation dynamometer. Following each test, the engines were disassembled, the deposits on the combustion chamber side of the valves were removed, and the intake valves were weighed. The weight obtained was compared to the weight of the valves before the test, with the difference being the total valve deposit weight. The average deposit weight per 25 valve (the sum of the deposit weights divided by the number of valves) is shown in Table 1 below.

Table 1

30	Run No.	Starting alcohol	moles of butylene oxide	Conc. ppm	mg/valve base fuel	mg/valve w/additive
35	A	Exxal 26	4	400	340	32
	B	Exxal 26	0	400	340	58
	C	Exxal 26	4	300	235	27
	D	Exxal 26	8	300	235	35
	E	Exxal 20	8	200	235	18
	F	Exxal 20	8	300	235	5
	G	Exxal 20	8	200	235	16
	H	Exxal 20	8	200	340	27
	I	Exxal 20	0	300	235	51

The data in Table 1 show that the most effective deposit control is obtained when the moles of butylene oxide ranges from 4 to 8.

45 Example 3 - Performance of Highly Branched Guerbet Ether Mono Amines Derived from Propylene Oxide

Analogues were made as described in Example 1 using propylene oxide instead of butylene oxide. A commercial unleaded 93 octane base gasoline containing these additives was then tested by running a BMW 325 50 for 100 hours on a standard mileage accumulation dynamometer. Following each test, the engines were disassembled and the deposit weights were quantified as in Example 1. The average deposit weight per valve obtained is summarized in Table 2 below.

Table 2

Starting alcohol	moles of propylene oxide	Conc. ppm	mg/valve base fuel	mg/valve w/additive
Exxal 26 (1)	0	400	340	58
Exxal 26	4	400	340	126
Exxal 26	8	400	340	154

10 (1) Run B from Table 1.

15 A comparison of the data in Tables 1 and 2 show that butylene oxide provides significantly improved detergent performance over propylene oxide analogues.

Example 4 - Performance of Linear, Non-Highly Branched, and Highly Branched Guerbet Ether Mono Amines

20 To further demonstrate the enhanced performance of highly branched Guerbet ether mono amines, analogues were made as described in Example 1 using other starting alcohols that contain 20 carbon atoms. These derivatives were all made with eight moles of butylene oxide. The results obtained when tested in a commercial unleaded 93 octane base gasoline are summarized in Table 3 below.

Table 3

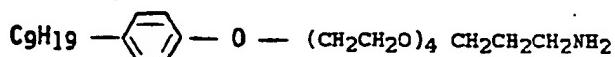
Starting alcohol	moles of buty- lene oxide	Conc. ppm	mg/valve base fuel	mg/valve w/additive
linear 20 carbon alcohol	8	200	235	120 (1)
20 carbon Guerbet alcohol from linear decyl alcohol (non- highly branched)	8	200	235	85
Exxal 20 (highly branched)	8	200	235	16

40 (1) An average of two separate runs (77 and 162).

45 The data in Table 3 show that derivatives made from highly branched Guerbet alcohols provide significantly enhanced performance in controlling intake valve deposits.

Example 5 - Performance of Ethoxylated Nonyl Phenol Mono Amine

50 A branched nonyl phenol derivative made with four moles of ethylene oxide and having the following structure



was prepared as described in Example 1. This additive (which is claimed in US Patent 3,440,029 as a deicer in carbureted vehicles) was tested in a commercial unleaded 93 octane base gasoline having a basal deposit

level of 235 mg/valv (using the above described BMW 325 test) when no additive was present. The gasoline was then blended with 200 parts per million by weight of the ethoxylated nonyl phenol mono amine and tested in the BMW 325 as described above. The additive containing fuel produced 610 mg/valve of deposit, which is significantly higher than the base fuel. Thus, some additives that are effective deicers can significantly increase intake valve deposits.

Example 6 - Performance of Highly Branched Guerbet Ether Mono Amines in Cleaning Electronic Port Fuel Injectors

Exxal 20 was reacted with 8 moles of butylene oxide and acrylonitrile as described in Example 1 to produce the Guerbet ether mono amine used in Run Nos. E-H in Table 1 of Example 1. This additive was tested in a 5 liter 8 cylinder electronic port fuel injected engine which had been removed from a Chevrolet Camaro. The engine was operated for 40 cycles using a commercial unleaded 87 octane base gasoline according to the procedure given in Table 4 below for a single cycle.

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Table 4

Time minutes	Engine Speed, rpm	Engine Load, ft. lb.	Coolant Jacket temperature, °F
10	2000	150	220
25	45	0	250

At the end of 40 cycles, the injectors were removed from the engine and the degree of injector fouling was quantified by measuring the amount of fuel which passed through the injectors per unit time. The percent fouling for the eight fuel injectors is given in Table 5.

35

Table 5

	<u>Injector No.</u>	<u>% Fouled</u>
40	1	3.9
	2	10.5
	3	23.3
	4	7.5
	5	8.9
45	6	6.9
	7	7.5
	8	6.1

The fouled injectors were then replaced in the engine and the engine was run on unleaded 87 octane gasoline containing 300 parts per million by weight of the above mentioned Guerbet ether mono amine (without other additives). After 25 additional test cycles, the injectors were again removed and the level of fouling quantified by measuring the fluid throughput. The level of fouling for the eight injectors is summarized in Table 6 below.

55

Table 6

	<u>Injector No.</u>	<u>% Fouled</u>
	1	0.0
	2	4.5
	3	2.6
10	4	4.3
	5	2.3
	6	1.6
	7	0.7
	8	0.9

15

A comparison of the data in Tables 5 and 6 shows that all eight fuel injectors were cleaned-up by fuel containing the highly branched Guerbet ether mono amines of this invention.

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Claims

1. A distillate fuel composition comprising (a) a gasoline, and (b) an alkyl ether mono amine having the formula:



where

R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and

x is from 0 to 30.

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2. A distillate fuel composition as claimed in claim 1 wherein R has the formula:



where R_1 is an alkyl group containing from 6 to 20 atoms, R_2 is an alkyl group containing from 4 to 18 carbon atoms, and R_1 and R_2 together contain at least four substituent methyl groups.

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3. A distillate fuel composition as claimed in claim 1 or 2 wherein R is derived from a Guerbet alcohol.

4. A distillate fuel composition as claimed in any preceding claim wherein R contains from 16 to 30 carbon atoms, preferably about 20 carbon atoms.

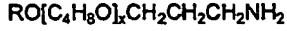
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5. A distillate fuel composition as claimed in any preceding claim wherein x is from 1 to 20, preferably from 4 to 8.

6. A distillate fuel composition as claimed in any preceding claim wherein from about 50 to about 2000 ppm of the alkyl ether mono amine is present in the fuel.

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7. Use of an alkyl ether mono amine having the formula:



where

R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and

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x is from 0 to 30

as a gasoline fuel additive for reducing the formation of intake valve deposits in an internal combustion engine.

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8. The use as claimed in claim 7 wherein the internal combustion engine has multipart electronically controlled fuel injectors.

9. The use as claimed in claim 7 or 8 wherein R and x of the alkyl ether mono amine are as defined in any of claims 2 to 5.

10. A fuel concentrate comprising:
(a) from about 10 to about 70 weight percent of an alkyl ether mono amine having the formula:
$$RO[C_4H_8O]_xCH_2CH_2CH_2NH_2$$
where
5 R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and
x is from 0 to 30, and
(b) from about 30 to about 90 weight percent of a hydrocarbon solvent, an alcohol solvent or a mixture thereof which boils in the range of from about 150° to about 400°F.
- 10 11. A fuel concentrate as claimed in claim 10 wherein R and x of the alkyl ether mono amine are as defined in any of claims 2 to 5.
12. An alkyl ether mono amine having the formula:
$$RO[C_4H_8O]_xCH_2CH_2CH_2NH_2$$
where
15 R is a highly branched alkyl group containing from 12 to 40 carbon atoms; and
x is from 1 to 30.
13. An alkyl ether mono amine as claimed in claim 12 wherein R and x are as defined in any of claims 2 to 5.
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European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 30 2385

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. CL.5)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	EP-A-0 353 713 (KAO) * Whole document * ---	1-13	C 10 L 1/22 C 07 C 217/08
X	GB-A-2 177 719 (KAO) * Claims 1,4; page 9, line 31 *	10-13	
A	---	1-9	
D, X	EP-A-0 310 875 (BASF) * Whole document * -----	1,4-13	
TECHNICAL FIELDS SEARCHED (Int. CL.5)			
C 10 L C 07 C			
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	23-05-1991	DE LA MORINERIE B.M.S.B.	
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